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54 Bitumenous compositions.

57 Bitumen modified with a modifier comprising a defined elastomeric component and a polyolefin component containing polyethylene and polypropylene has improved high temperatures and low temperature properties which make it especially suitable for roofing applications. The elastomeric component is an ethylene propylene copolymer having a broad compositional distribution with regard to its ethylene content.

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Bitumenous Compositions

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This invention relates to bitumenous compositions having improved high temperature and low temperature properties, particularly but not exclusively such compositions which may be used in roofing applications.

10

Bitumen is frequently used in roof coverings, often in the form of layers of fabric which are impregnated with a soft bitumen and coated with a harder bitumen which may be a blown bitumen containing mineral fillers.

15

However, these materials suffer from the problem that they cannot withstand prolonged outdoor exposure because temperature variations can lead to cracks in the harder bitumen layers and to non-recoverable deformations due to creep or flow of the viscous materials. To overcome this several layers for example five layers of bitumenous roof covering are often used, but this inevitably leads to high laying and maintenance costs.

20

Attempts have been made to produce bitumenous felts and suggestions have been made to improve the properties of the felts by modifying the bitumen with certain polymers.

25

As yet however it has not been possible to obtain bitumenous compositions which are completely satisfactory and readily produced, and which form commercially acceptable roofing materials having the desired properties. Ideally the modifier for the bitumen should

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1 be easily incorporated therein from particulate form to
yield a homogenous composition which remains homogenous
on storage, and should have a viscosity which permits
use in normal roofing material manufacturing equipment.
5 Thus, it is desirable that the roofing material obtained
should have substantially no creep below 100°C, should
have adequate flexibility at low temperatures, for
example, down to -10°C and as low a permanent set as
possible after 25% elongation. In addition these
10 properties should preferably be retained after 6 months
at 70°C, or 1 month at 80°C. In addition to thermal
stability it is desirable that the material should
stand up well to UV radiation in accordance with
the xenon test. This particular feature is however not of
15 great importance since although it is now common in
certain cases to leave the bitumenous roofing materials
exposed to sunlight, whereas hitherto it has been
conventional to coat the materials with slate or
chippings which would protect the base material from UV
20 radiation, the base material is of course black and so
UV attack is in any case only superficial.

It has been proposed to modify the properties of
bitumen by blending with styrene-butadiene block
copolymers. Although this generally improves the
25 properties of the bitumen immediately after blending
such as its low temperature creep resistance, flexi-
bility and elastic recovery, it may have an adverse

1 effect on the heat ageing and UV radiation resistance
properties of roofing materials formed from the blend.
Furthermore, styrene-butadiene copolymers can only be
used in conjunction with certain specific bitumens,
5 otherwise they are subject to a high mixing rejection
rate, that is, separation out from the bitumen. It is
also known to include atactic polypropylene to form a
bitumen blend suitable for use on flat roofing applica-
tions; however the balance of properties obtained with
10 such blends are generally not optimal.

It has been proposed in US Patent 3 669 918 (Dow
Chemical Company) to include ethylene propylene copolymer
rubbers into bitumen for roofing applications, but with
no teaching as to the compositional distribution of
15 such rubbers. We have found however that such rubbers
may only be incorporated directly into bitumen if they
have low molecular weight, since higher molecular
weight polymers are impossible to disperse in the
bitumen when using normal blending conditions. Moreover,
20 incorporation of ethylene propylene rubbers alone into
bitumen has been found to produce compositions having
softening points below 100°C, whereas it is highly
desirable for the softening point to be greater than
100°C or even 120°C.

25 The incorporation into bitumen of ethylene propylene
rubbers of high ethylene content, which are usually not
readily usable with the bitumen, has been achieved

1 according to GB 1 427 307 (SIPLAST) by using a poly-
olefin as a solid dispersion aid. Thus, the rubber and
polyolefin are blended and granulated, and this granular
premixture is then mixed at 180 to 250°C with the
5 bitumen to give the required dispersion. Similarly,
GB 1 400 145 (Briggs Amasco Limited) discloses
bitumenous compositions comprising a mixture of bitumen
with ethylene propylene ethylidene norbornene terpolymer
and polyethylene in a minor amount compared with the
10 rubber. However, neither of these documents teaches
the use of rubbers having a broad compositional distri-
bution with regard to the ethylene content.

GB 1 519 508 (HULS) discloses the manufacture of
homogenous thermoplastic compositions in ribbon or
15 granule form, which comprise blends of defined poly-
olefin rubber, optionally additionally a polyolefin,
and bitumen. The rubber may be an ethylene propylene
rubber which is a sequence polymer, that is a copolymer
containing ethylene or an alpha-olefin in addition to a
20 statistical distribution of the monomers in the polymer
chain in the form of chain sections of varying length
(sequences). This gives the polymer good raw strength
(at least 20 kp/cm² at 25°C) and hence permits
manufacture in comminuted form which is advantageous
25 for the blending process. Again, though, there is no
teaching to use rubbers of broad compositional distri-
bution with regard to the ethylene content.

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1 US 4 110 414 (Goodrich) discloses mouldable thermo-
plastic polymer blends of high tensile strength prepared
by mixing low density polyethylenes and high molecular
weight ethylene propylene copolymers under conditions
5 of heat and shear. The EP copolymer has a methylene
run index, i.e., a measure of the ratio of methylene
units in runs of seven or more to the sum of the runs
of three methylene units plus the runs of two methylene
units, of 20 or above, but there is no suggestion here
10 that such blends might be useful for modifying bitumen.

It has now surprisingly been found that bitumen may be
advantageously modified, particularly but not exclusively
for roofing applications, by a combination of a poly-
olefin component and an elastomeric component of
15 specific characteristics.

According to the present invention there is provided a
bitumenous composition which comprises bitumen and,
dispersed therein in an amount sufficient to modify the
bitumen to improve its low temperature and high tempera-
20 ture properties, particularly its suitability for
roofing applications, a modifier comprising a polyolefin
component containing polyethylene and polypropylene,
and an elastomeric component characterised in that the
elastomeric component comprises an ethylene-propylene
25 copolymer having a broad compositional distribution with regard to
its ethylene content. Such bitumenous compositions are also termed
"polymer asphalt blends" (PAB) herein.

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1 According to a further aspect of the invention there is
provided the use of a modifier composition as defined
above as an additive for bitumen.

By ethylene-propylene copolymers (EPR) having a broad
5 compositional distribution (BCD) there is meant EPR
having individual polymeric chains with respective
ethylene contents which are broadly distributed about
the average ethylene content of the copolymer. Typically
EPRs with broad compositional distribution include
10 polymeric chains of ethylene content at least 5,
preferably at least 10, weight percent absolute greater
than the average ethylene content of the copolymer, and
polymeric chains of ethylene content at least 5,
preferably at least 10, weight percent absolute less
15 than said average ethylene content. Preferably
for the purposes of this invention the ethylene content
of the individual polymer chains includes values of
from 5 to 25, more preferably 10 to 25, wt% absolute
above and 5 to 25, more preferably 10 to 25 wt% absolute
20 below the ethylene content of all the polymer chains
(depending on the average ethylene content). Thus, for
a BCD-EPR of ethylene content 40%, individual chains
could be extracted which have ethylene contents as low
as 15 wt% to as high as 65 wt% for the broadest
25 compositional distribution.

The term ethylene propylene copolymer (EPR) as used
herein means an elastomeric polymer formed from at

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1 least two monomers, one of which must be ethylene and
one propylene. Thus, it includes EPM rubbers, and also
unsaturated rubbers where a minor proportion of one or
more polyenes such as the endomethylenes, 5-ethylidene
5 norbornene, 5-methylene norbornene and dicyclopentadiene,
or a non conjugated aliphatic diene such as 1,4-
hexadiene is included in the polymeric chains, commonly
termed EPDM rubbers.

The compositional distribution of an EPR is characterised
10 by the variation in ethylene to propylene ratio among
the chains and within the chains. For example if all
the chains have the same composition and the chains
have that composition from end-to-end then the material
is said to have a narrow compositional distribution.
15 If chains, one versus another, vary greatly in composition
then the compositional distribution is said to be
broad that is the individual polymer chains do not all
have the same % content of ethylene monomeric units,
but together span a range of ethylene contents.

20 The compositional distribution of an EP copolymer may
be determined for example by using extraction techniques,
solvent fractionation, or gel permeation chromatography
to split the copolymer into its various molecular
weight fractions. The ethylene content of these
25 fractions is then measured by a standard method such as
infra-red absorption or nuclear magnetic resonance.
Thus according to one fractionation technique the

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1 copolymer is dissolved in a solvent such as hexane, and
aliquots of a precipitant such as an alcohol are
progressively added. High molecular weight fractions
are precipitated first, with lower molecular weight
5 fractions being deposited as the alcohol concentration increases. According to another technique the
solid copolymer is deposited on glass beads, and then
progressively eluted therefrom by treatment with
amounts of progressively more powerful solvent, for
10 example alcohol/hexane mixtures with increasing hexane
concentration, or by the same strength solvent at
increasing temperatures. Here, the low molecular
weight fractions are eluted first, followed by higher
molecular weight fractions. It has been found that
15 with EPRs having a low average ethylene content, e.g.
approx. 40 wt%, the longer chain (higher molecular
weight) fractions tend to have a higher ethylene
content than the shorter chain (lower molecular weight)
fractions. However this is not necessarily the case
20 with broad compositional distribution EP copolymers
with higher average ethylene contents.

The compositional distribution of the copolymer may be
defined, as above, in terms of the extremes of composition versus the average ethylene content; or
25 alternatively by determining the entire range of
composition among the chains.

1 It has been found that modified bitumens in accordance
with the invention yield much better properties (for
example low temperature and elastic). Thus at low
temperatures close to glass transition temperature the
5 flexibility of the material is increased whilst at high
temperatures e.g. glass transition temperature + 130
degrees C, the flow is decreased i.e. softening point
is increased. By high temperatures as mentioned
hereinbefore there is meant the sort of temperatures to
10 which roofing felts incorporating the PAB may be
routinely subjected in use at various locations through-
out the world, or to which PABs are subjected during
testing for suitability for such applications. For
example these high temperatures may be in the range
15 70-130°C, particularly 80-120°C and especially 100-120°C.
It is emphasised here, though, that the PABs according
to the invention are not limited in their use to
roofing applications, but may also be used for example
in pipe-wrapping applications. By low temperatures
20 as mentioned hereinbefore there is meant the sort of
temperatures to which for example rolls of roofing felt
incorporating the PAB may be subjected prior to un-
rolling such rolls for application to a roof surface at
various locations worldwide, or to which PABs are
25 subjected for testing their suitability for such
applications. For example these low temperatures may
be in the range 0 to -30°C, particularly -15 to -25°C.

- 1 For roofing applications, such modified bitumens
compare well with bitumen treated with conventional
modifying systems such as those wherein the EP copolymers
have a narrow compositional distribution or wherein the
5 EPR is not in combination with the polyolefin component.
It is speculated that this improvement derives from the
interaction of the broad compositional distribution
polymer chains with the polyolefin components of the
modifier system and the bitumen.
- 10 The use of BCD-EPRs in bitumen (asphalt) modifier
systems according to the invention has been found to be
particularly advantageous when the copolymer has a low
average ethylene content, for example less than 50% or
53% by weight, particularly from 40 to 45 wt%. In
15 general, the lower the ethylene content of an EPR, the
lower is its crystallinity. It is known that bitumen
modified with EPR of high crystallinity (high ethylene
content) has an improved mechanical strength, but by

1 virtue of this high ethylene content such EP copolymers
are considerably more difficult to incorporate into the
bitumen than low ethylene content EPRs. According to
the invention modifier systems including EP copolymers
5 of low average ethylene content but broad compositional
distribution not only give improved properties to the
bitumenous composition, but also are more readily
incorporated into the bitumen by the techniques common
in the art. It is speculated that low ethylene content,
10 broad compositional distribution EP copolymers have,
despite their low average ethylene content, an increased
crystallinity because they include polymeric chain
segments containing ethylene in a proportion which is
at least 5 wt% absolute greater than the average.
15 Thus, certain copolymer chain segments will effectively
contain blocks of "polyethylene" which interact with
each other and with the polyethylene in the modified
bitumen. The lower ethylene content chain segments
interact with the polypropylene and the bitumen. With
20 normal narrow compositional distribution EP copolymers
(NCD-EP) much higher ethylene contents are required

1 to yield such interaction with polyethylene. Moreover
there are fewer low ethylene chain segments in the high
ethylene NCD-EP copolymers to interact with the bitumen
and polypropylene. Of course in accordance with the
5 invention BCD-EPRs with relatively high ethylene
contents, e.g., 50 to 70 or even 80% may be used, but
here the advantages of the BCD are less evident by
virtue of the inherent high crystallinity of these materials.

According to another aspect of the invention there is
10 provided a method of producing bitumenous compositions
as hereinbefore defined which comprises first forming a
modifier masterbatch of the BCD-EPR and the polyolefin
component containing polyethylene and polypropylene,
and subsequently incorporating the masterbatch into the
15 bitumen.

By way of example the masterbatch may be prepared by
incorporating all the ingredients in an internal mixer
e.g. Banbury type, and mixing for about 3 minutes at
flux temperature e.g. about 140°C. Subsequently the
20 mixture is pelletised using a single screw extruder,
and the pellets dusted with talc to prevent agglomeration.
This method is useful for laboratory scale work. For
more economical bulk scale work, mixing and pelletisation
may be performed for example in a high intensity dual
25 screw continuous mixer, such as a Werner-Pfleiderer or
a Farrel, with a die temperature of about 180°C.

1 Any form of bitumen may be used provided it is compatible
with the modifier. For example the bitumen may be
Safaniya 180/220, which is a non-blown (non-oxidised)
straight bitumen of moderate aromaticity from Saudi
5 Arabia.

It has been found that such modifiers may be incorpo-
rated into the bitumen using conventional blending
equipment to give a homogeneous blend which is generally
stable on storage and of a viscosity that may be
10 processed under normal roofing material manufacturing
conditions. Thus yet another aspect of the invention
provides for the use of such bitumenous compositions as
roofing materials, which are generally found to have
desirable physical properties which are retained after
15 heat ageing.

By way of example, the modifier may be incorporated
into the bitumen under laboratory conditions by heating
the bitumen to 200-205°C in a rapidly (1000 rpm)
stirred glass reactor and adding modifier in pellet
20 form over 15 minutes without vortex formation, stirring
being continued for about 1 hour after the addition.
Dispersion quality may be checked visually (film test,
or preferably using a microscope). Alternatively paddle
mixing at e.g. 60 rpm for about 4 hour may be employed.
25 On a bulk scale an industrial paddle mixer at e.g. 20
rpm may be used, with addition of modifier over e.g. 1

1 hour and further mixing at reduced temperature e.g. 180-
190°C., optionally with addition of filler, for about 1 hour.

The modifier system is preferably incorporated in the
bitumen in a proportion of from 5 to 20% by weight
5 based on the composition as a whole, such a proportion
having been found to give optimum end product physical
properties in conjunction with commercial acceptability.
More preferred is the range 9 to 16 wt% and particularly
the range 12 to 16 wt%. Such compositions generally
10 have been found to have good high temperature properties,
e.g., softening points in excess of 100°C, which is a
requirement for roofing applications (thought to derive
in part from the polyethylene content of the polyolefin
component); and good low temperature properties, e.g.,
15 brittle failure only at -10°C or below (thought to
derive in part from the polypropylene content of the
polyolefin component).

The polyethylene is preferably high density polyethylene
(HDPE), and the polypropylene is preferably atactic
20 polypropylene (APP), with the ratio between these two
in the polyolefin component depending to a great extent
on the properties required of the bituminous composition.
For example APP tends to depress softening point but
improve low temperature characteristics, whereas HDPE
25 tends to increase the softening point for example to
120-130°C which is highly desirable for roofing applica-
tions. However HDPE may be detrimental to elasticity

1 by virtue of its high ⁻¹⁵ crystallinity. From the above it
may be considered that the APP functions as a plasticizer
for the rubber, and in this regard it functions even
for EPRs which do not have a broad compositional
5 distribution.

As mentioned, the composition of the modifier may vary
in dependence on the desired end properties. Preferably
the modifier will comprise from 40 to 60 wt% of the
elastomeric component, or indeed even greater amounts
10 of BCD-EPR, for example from 50 to 75%, with the
balance being polyolefin component. In this polyolefin
component the ratio of polyethylene to polypropylene is
preferably in the range 40:60 to 60:40 by weight, more
preferably in the ratio 1:1. A particularly useful
15 modifier system contains BCD-EPR:APP:HDPE in the weight
ratio 50:25:25. The modifiers may also contain additives
such as stabilizers and antioxidants. Preferably
reinforcing fillers such as carbon black or silica are
not used since although they can improve the softening
20 point of the bitumenous composition, their presence
tends to increase brittle failure at low temperatures.

The BCD-EPRs which have been found to be useful in
bitumenous compositions according to the invention
may be produced by conventional copolymerisation
25 techniques adapted to yield the BCD product by control
of process conditions as is well understood in the art.
For example the degree of agitation in the copoly-

1 merisation reactor can be adjusted to give the desired
product. Alternatively, of the numerous copolymerisation
catalyst systems that are known, the various combinations
of catalyst and cocatalyst can be adjusted by relatively
5 simple testing methods to yield EPRs having the compositional
distribution of any sufficient breadth for the proposed end
use of the bitumenous composition.

The bitumenous compositions of the present invention may
be processed into roofing materials by standard proces-
10 sing techniques. They may be used to impregnate felts
and as intermediate layers in the traditional multilayer
roofing materials; however they are primarily intended
for use in the preparation of single or double layer
roofing materials. In either circumstance it has been
15 found that the use of the bitumen compositions of the
present invention permits the fabrication of roofing
materials of improved viscoelastic properties.
Softening point, low temperature flexibility and ageing
characteristics are also generally improved.

20 The following Examples illustrate the invention.

Example 1

A modifier masterbatch was produced by blending together
a BCD-EPR, APP and HDPE in a ratio by weight of 50:25:25.
The EPR had an average ethylene content of 42% by
25 weight, and a broad compositional distribution (as
hereinbefore defined) by virtue of the catalyst system
employed in its production. Its density was 0.86 g/cc

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1 (23°C); $M_L(1+8)$ value 33 at 100°C inherent viscosity
3.34 (in decalin at 135°C); and molecular weight
characteristics (by GPC in 1,2,4 trichlorobenzene at
135°C) $M_n = 6300$, $M_w = 296,000$, $M_w/M_n = 47$, broad MWD.
5 The APP was AMOCO grade 115-601 A of density 0.85 g/cc
at 23°C, Brookfield viscosity range 200-1200 cps at
190°C, and ash content 0.05 wt.% max.
The HDPE was HD 6950 (ESSO), a narrow molecular
weight distribution product of density 0.963 g/cc
10 (23°C, ASTM D 1505); and melt index 7 g/10 min (ASTM D
1238, 2.16 kg load).

Blending was by incorporating all the ingredients in a
Banbury internal mixer and mixing at flux temperature
(about 140°C) for 3 minutes. Thereafter the blend was
15 pelletised in a single screw extruder and the pellets
dusted with talc to prevent agglomeration.

To yield a bitumenous composition containing 12 wt%
modifier and 88 wt% bitumen, the modifier was then
incorporated into a 200 penetration straight bitumen of
20 Safaniya origin. Thus the bitumen was
heated to 200-205°C in a rapidly stirred glass reactor
and masterbatch pellets were added over a 15 minute
period, without vortex formation (but with dimples
being observed at the bitumen/air interface). After the
25 addition mixing was continued for 1 hour.

The composition was tested for its suitability for
roofing applications by the following methods.

- 1 (a) Softening point: by the "Ring and Ball" method,
ASTM D-36, expressed in °C.
- (b) Foldability: by a non-standard method, a strip
of the bitumenous composition (2x30x100m) is
5 conditioned at a constant temperature for 1
hour and then folded (over a period of 5
seconds) around a mandrel of 10mm diameter.

If the sample does not exhibit surface micro-
cracks, the procedure is repeated at a lower
10 temperature (reduced by 5 degrees C stepwise)
until such time that microcracks do appear
onfolding, the temperature of first cracking
being considered as the foldability value.

Results of the tests showed a high softening point of
15 119°C and a low foldability value of -10°C, these being
excellent values for roofing applications.

Example 2

The procedure of Example 1 was repeated but using a
BCD-EPR of average ethylene content 65 wt%. Test
20 results gave a softening point of 116°C and a fold-
ability value of -10°C.

Comparison Example 3

Example 1 was repeated, but using an EPR which although
having an ethylene content of 42%, had a narrow composi-
25 tional distribution. Test results gave values of 84°C
and -5°C respectively for softening point and foldability.

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- 1 Such values are not acceptable for roofing applications where the product will be subjected to extremes of hot and cold weather.

Comparison Example 4

- 5 Example 1 was repeated, using an EPR with an ethylene content of 65%. The EPR had the same Mooney viscosity as that of Example 1, but an extremely narrow compositional distribution. Moreover it had a narrow MWD ($M_w/M_n = 2$). The crystallinity of this rubber was
- 10 so high that it was found to be extremely difficult to produce the modifier in the Banbury mixer. Thus a much longer (and hence less economical) mixing time was required to form the modifier blend. Test results gave values of 119°C and -10°C for softening point and
- 15 foldability respectively which of course compares well with Example 1, but for a much higher ethylene content of the rubber, and a much less convenient and uneconomical blending procedure.

1. A bitumenous composition which comprises bitumen and, dispersed therein in an amount sufficient to modify the bitumen to improve its low temperature and high temperature properties, a modifier comprising a polyolefin component containing polyethylene and polypropylene, and an elastomeric component, characterised in that the elastomeric component comprises an ethylene-propylene copolymer having a broad compositional distribution with regard to its ethylene content.
2. A composition according to claim 1, wherein the copolymer includes polymeric chains of ethylene content at least 5 wt% absolute greater than the average ethylene content of the copolymer, and polymeric chains of ethylene content at least 5 wt% absolute less than said average.
3. A composition according to claim 2, wherein the copolymer includes polymeric chains of ethylene content from 5 to 25 wt% absolute greater than the average and from from 5 to 25 wt% absolute less than said average.
4. A composition according to claim 1, 2 or 3, wherein the copolymer has an average ethylene content of less than 53 wt%.

5. A composition according to claim 4, wherein the copolymer has an average ethylene content of from 40 to 45 wt%.
6. A composition according to any one of the preceding claims, wherein the copolymer comprises EPM rubber.
7. A composition according to any one of the preceding claims, wherein the copolymer comprises EPDM rubber.
8. A composition according to any one of the preceding claims, wherein the polyethylene is high density polyethylene and/or the polypropylene is atactic polypropylene.
9. A composition according to any one of the preceding claims, which comprises from 5 to 20 wt% modifier, based on the composition as a whole.
10. A composition according to claim 9, which comprises from 12 to 16 wt% modifier.
11. A composition according to any one of the preceding claims, wherein the modifier comprises from 40 to 60 wt% of elastomeric component, based on the modifier as a whole.
12. A composition according to any one of the preceding claims, wherein the polyolefin component comprises polyethylene and polypropylene in a ratio of from 40:60 to 60:40 by weight.

13. A composition according to claim 1, which comprises from 12 to 16 wt% of a modifier comprising copolymer, atactic polypropylene and polyethylene in a weight ratio of 50:25:25, said copolymer having an average ethylene content of about 40 wt% and containing individual chains with ethylene contents of about 15 to 35 wt% and about 45 to 65 wt%, and from 84 to 88 wt% of a 180/220 penetration bitumen.
14. A method of producing a bitumenous composition according to claim 1, characterised in that the polyethylene, polypropylene and ethylene-propylene copolymer are blended together to form the modifier, and said modifier is then mechanically dispersed as a masterbatch in the bitumen.
15. A bitumenous composition according to any one of claims 1 to 13, or when produced by the method according to claim 14, when in the form of a roofing material.
16. A method of protecting a roof which comprises applying thereto a bitumenous composition according to claim 15.
17. The use as an additive for bitumen of a modifier composition comprising a polyolefin component containing polyethylene and polypropylene, and as elastomeric component, an ethylene-propylene copolymer having a broad compositional distribution with regard to its ethylene content.



European Patent
Office

EUROPEAN SEARCH REPORT

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Application number

EP 84 30 4027

| DOCUMENTS CONSIDERED TO BE RELEVANT | | | |
|---|--|--|---|
| Category | Citation of document with indication, where appropriate, of relevant passages | Relevant to claim | CLASSIFICATION OF THE APPLICATION (Int. Cl. *) |
| A | FR-A-2 228 817 (SOCIETE NOUVELLE SIPLAST) * Page 2, lines 12-28; page 7, claims 3,4 * & GB - A - 1 427 307 (Cat. D) | | C 08 L 95/00 C 08 L 23/02 // C 08 L 23/16 C 08 L 23/06 C 08 L 23/12 |
| A,D | GB-A-1 400 145 (BRIGGS AMASCO) | | |
| A | US-A-4 368 228 (ROMOLO GORGATI) | | |
| A | DE-A-2 219 147 (PHOENIX GUMMIWERKE) | | |
| A | US-A-3 963 659 (GERHARD BINDER) | | |
| | | | TECHNICAL FIELDS SEARCHED (Int. Cl. *) |
| | | | C 08 L |
| The present search report has been drawn up for all claims | | | |
| Place of search THE HAGUE | | Date of completion of the search 05-10-1984 | Examiner GIRARD Y.A. |
| CATEGORY OF CITED DOCUMENTS | | | |
| X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document | | T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document | |